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A Germanium Detector with Optimized Compton Veto for High Sensitivity at Low Energy

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A Germanium Detector with Optimized Compton Veto for High Sensitivity at Low Energy

Final Report of the MPACT Optimized Compton Veto Detector Project (FTLL11MP0206)

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Abstract

We have built a prototype germanium detector with a Compton veto that is optimized for high sensitivity in the low-energy range around ~100 keV. It is specifically designed to address the problem to directly detect plutonium gamma emissions in spent nuclear fuel by non-destructive assay. This is not possible with current detectors due to the large low-energy background of Compton-scattered high-energy radiation from the fission products, whose gamma flux is at least 6 to 7 orders of magnitude higher than the Pu signal. Our instrument is designed to assess the feasibility to selectively suppress the background in the low-energy region around ~100 keV with the strongest Pu X-ray and gamma emissions lines. It employs a *thin* Ge detector with a large Compton veto *directly behind* it to suppress the background from forward-scattered radiation by anti-coincidence vetoing. This report summarizes the design considerations and the performance of the instrument.

Table of Contents

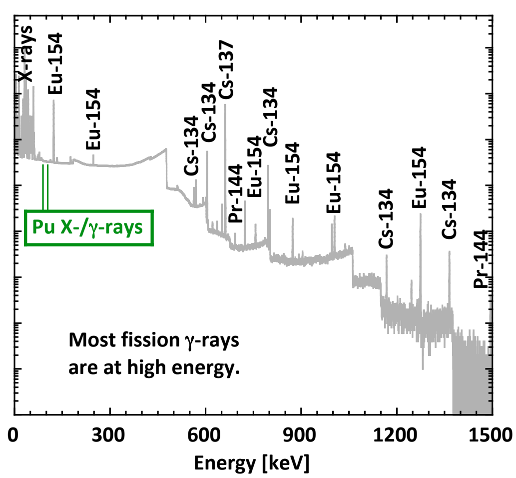
A Germanium Detector with Optimized Compton Veto for High Sensitivity at Low Energy

1. Introduction

The difficulty to directly detect and (ideally) quantify Pu in spent nuclear fuel by non-destructive assay is a significant challenge for nuclear safeguards. The problem arises from the large background at low energies due to Compton scattering of high-energy radiation from the fission products, whose gamma flux is at least 6 to 7 orders of magnitude higher than the Pu signal. In this FY11 project (work package FTLLMP0206), we have designed, simulated, built, tested and optimized a Ge detector with a Compton veto specifically optimized for high sensitivity at low energies in the presence of intense high-energy radiation. Here we summarize the design considerations and the performance of the instrument.

2. Spectrometer Design

The instrument exploits the fact that most gamma emissions from fission products have energies *well* above the Pu lines around ~100 keV (figure 1), so that the relevant background is mostly set by *forward* scattered radiation in the Ge detector. Sensitivity at low energies can therefore be increased with a *thin* Ge detector and a large Compton veto *directly behind* it. The Ge detector should be no thicker than required to absorb most of the low-energy radiation of interest to minimize the amount of Compton scattering by high-energy fission gammas. The Compton veto should be as large as possible and located directly *behind* the Ge detector, so that high energy gammas that are forward scattered in the Ge can be rejected with an anti-coincidence veto. To ensure that the low-energy background is in fact dominated by Compton interactions *inside* the Ge crystal rather than in the surrounding parts of the instrument or the laboratory, the source emissions should be collimated onto the Ge detector. Any mass in the volume defined by the opening angle of the collimator that is not part of an active detector volume should be minimized (figure 2). This includes the requirement to minimize the mass of the source itself.



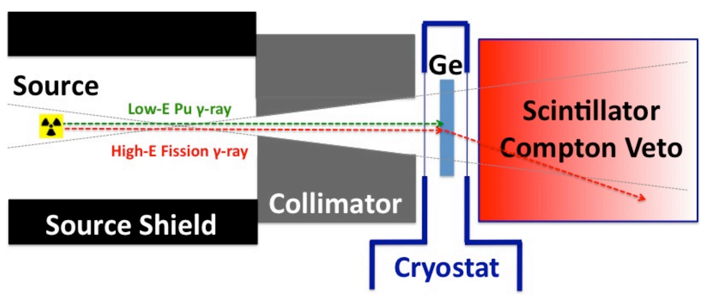
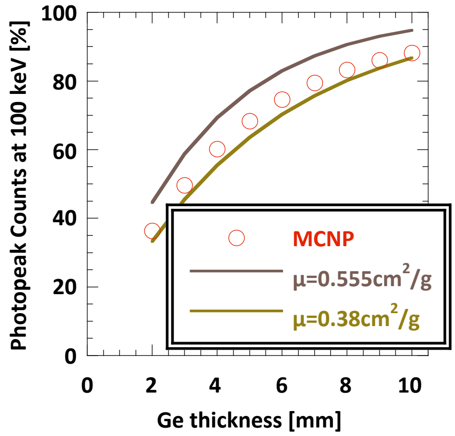


Figure 1 (left): MCNP simulations from a benchmarked spent fuel composition of a South Korean PWR reactor. Figure 2 (right): Schematic design of a Ge detector optimized for high sensitivity at low energy. The collimation angle (dotted grey line) defines the volume in which the (non-detector) mass must be minimized.

2.1. Germanium Detector

For best peak-to-background ratio, the Germanium detector should be no thicker than required to absorb the majority of the ~100 keV Pu gamma rays of interest, since further increases in thickness will no longer increase the signal significantly while adding to the Compton background. We have chosen a



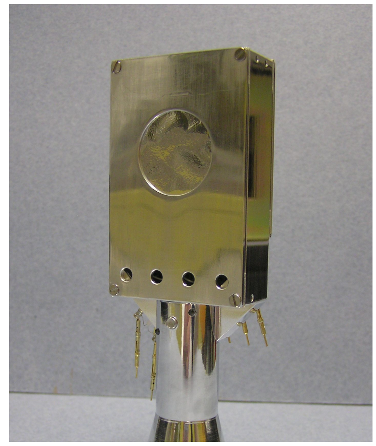
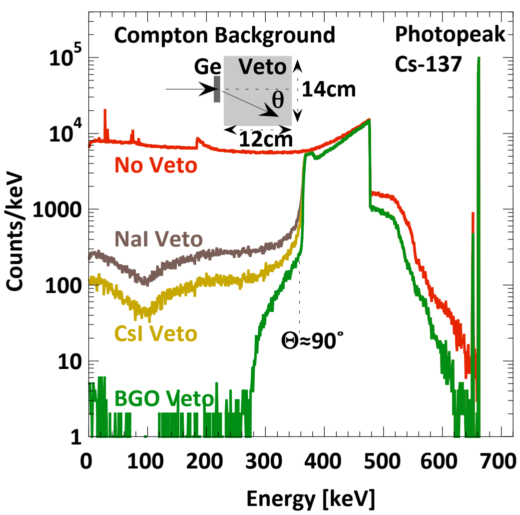


Figure 3 (left): Photopeak efficiency at 100 keV in Ge detectors with varying thickness. Figure 4 (right): Photograph of the detector holder optimized for minimum mass in the direct line of sight in front of *and* behind the Ge crystal.

Ge detector with a thickness of 8 mm that absorbs >80% of the Pu signals at ~100 keV (figure 3). The Ge crystal has a diameter of 35 mm and is only held at the edges to suppress Compton scattering in the support structure, with two 25-µm-thick Al windows to absorb thermal photons (figure 4). The cryostat is designed with flat surfaces close to the detector holder for close placement of the collimator and the vetoes. The only mass in the direct line of sight of the collimator consists of two Al vacuum windows in front and behind the detector, each 0.76 mm thick with a 4 cm diameter (cf. figure 2).

2.2. Scintillator Compton Veto

The scintillator should detect as many forward-scattered Compton photons as possible for best background suppression of the low-energy background. The design is therefore optimized with the biggest (figure 6) and highest-efficiency (figure 5) crystal that the budget can afford, subject to two constraints: For large volumes the event rate will exceed the scintillator's maximum count rate, in which



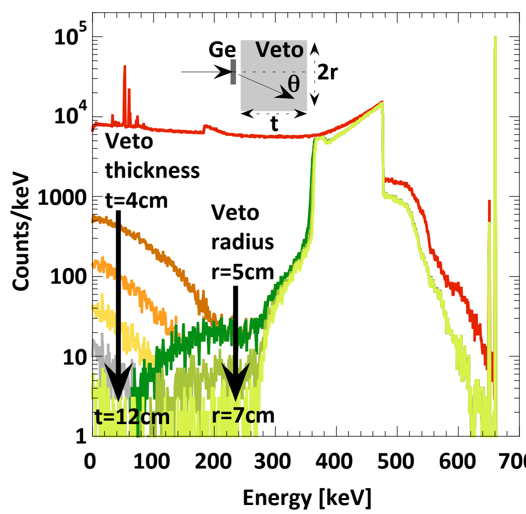
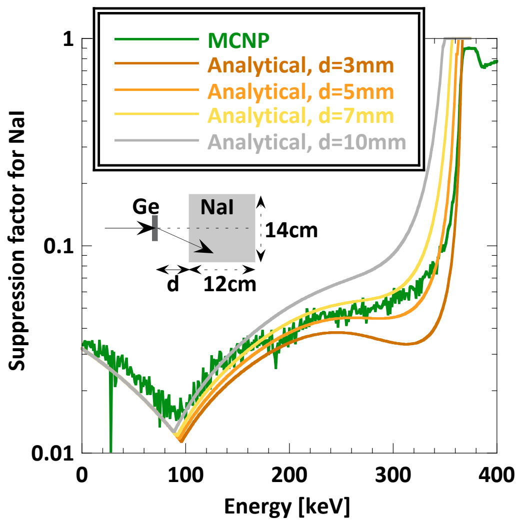


Figure 5 (left): MCNP simulations show how anti-coincidence vetoing selectively suppresses the Compton background in the low energy region of interest if the veto is located *behind* the Ge detector. The higher the quantum efficiency of the scintillator, the better the background suppression. Figure 6 (right): MCNP simulation of the background suppression vetoes of different sizes. In general, the bigger the better.

case it should be replaced by multiple scintillators to cover the required volume. Secondly, increases in veto efficiency will not reduce a background that it is set by scattering *outside* the Ge detector.

In general, the background suppression factor is given by the fraction of Compton photons that interact and leave a measureable signal in the vetoes. For simple axially symmetric geometries, the influence of geometrical changes, such as the distance between Ge detector and crystal, can therefore be estimated analytically (figure 7). Similarly, the penalty for certain unavoidable non-idealities such as the metal enclosure of a scintillator can be estimated from the fraction of Compton photons absorbed in it.

For this project we have used a cylindrical primary veto made from CsI with a diameter of 3.5" and a length of 6". The diameter is chosen so that it fits tightly into a secondary annular CsI veto with a 3.5" inner diameter from an earlier project that we had access to. GEANT4 simulations of the background suppression these two vetoes can provide in the ideal case of negligible scattering outside the Ge detector are shown in (figure 8). Possible future improvements for larger and more efficient vetoes are included for comparison.



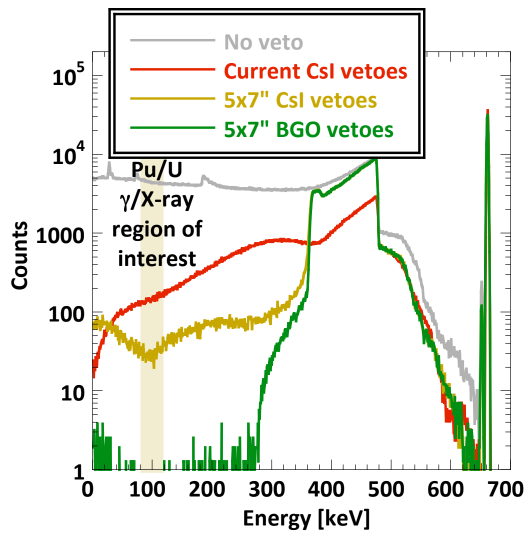


Figure 7: Comparison of the suppression factor between Monte Carlo simulations and analytical solutions for simple geometries. The maximum suppression corresponds to a scattering angle towards the corner of the veto. Figure 8: GEANT4 simulation of the background suppression for the two CsI vetoes used for this project (red).

2.3. Source Design

Scattering inside the radioactive source itself is one contribution to the background that cannot be rejected with the scintillator veto and that ultimately limits the attainable Compton suppression. It is therefore essential to keep the source itself and its protective cover as thin as possible, subject to radiation safety concerns. MCNP simulations show that the scattering contribution can be kept below 0.1% and above the 100 keV region of interest if the sources are sufficiently thin so that multiple in-source scattering is negligible (figure 9).

Our collaborators at Savannah River (work package FTSR11MP0213) have therefore diffused solutions of radioactive material into ~0.5 mm thick stainless steel planchets with a Bunsen burner. (0.1 mm planchets did buckle.) Since Cs-137 often dominates the fission background in spent fuels, SRNL fabricated test sources of 1) pure Cs-137, 2) Cs-137 with reactor-grade Pu, 3) spent fuel with additional reactor-grade Pu, and 4) spent fuel only (figure 10). The spent fuel from the Oak Ridge Research Reactor Fuel Element had been burned for ~1090 days in the core, and then cooled for ~8700 days. For safety, the planchets were subsequently encapsulated in 200 µm vinyl / 25 µm teflon (VF-81) protective tape.

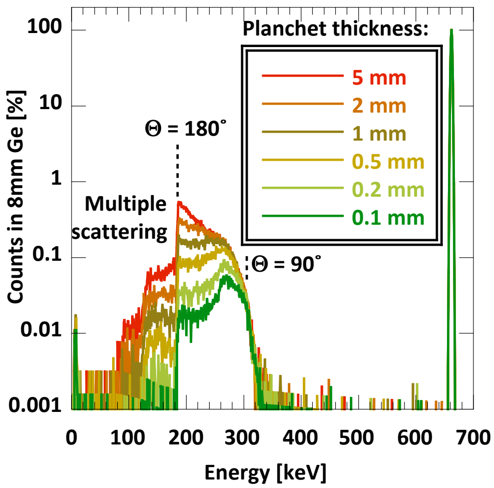
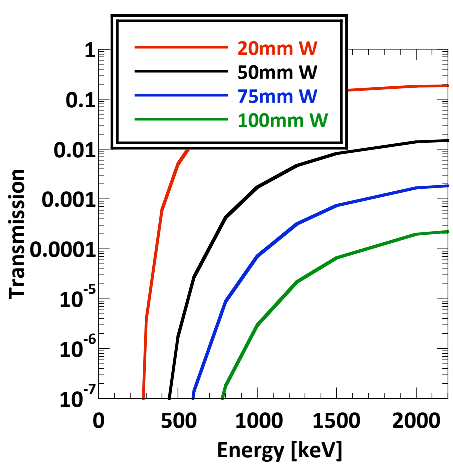


Figure 9 (left): MCNP simulation of the background contribution for Cs-137 sources in stainless steel planchets with different thickness. The angle refers to the scattering angle inside the source that causes background at the corresponding energy. Figure 10 (right): Set of four radioactive test sources from SRNL (David DiPrete, SRNL)

2.4. Collimator

The other unavoidable contribution to the Compton background is due to scattering in the collimator that is required to keep the fission gammas from scattering in the surrounding laboratory environment. We are using a 4" thick tungsten collimator with a straight 1.5" inner diameter that suppresses all lines from the fission products by at least 4 orders, and the strongest emissions <1 MeV from the Cs isotopes by at least 6 orders of magnitude (Figure 11). To reduce the small-angle scattering in the collimator, its opening should be modified in the future to be conically shaped, and the radioactive source should be placed such that no gammas can directly illuminate its inside wall (cf. figure 2). In this case, only gammas scattered close to the collimator entrance hole can reach the Ge detector, and they are mostly limited to scattering angles with sufficiently low energy loss that the background they cause is far from the ~100 keV region of interest, at least for the common fission gammas below 1300 keV where single scattering events dominate (figure 12).



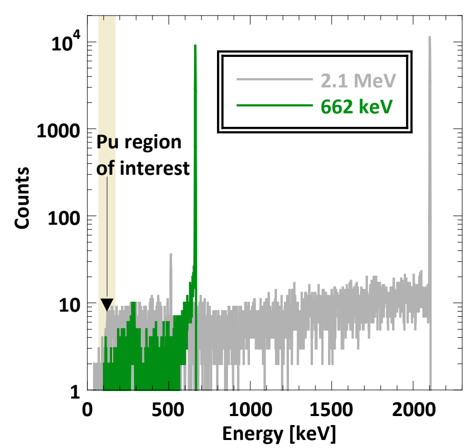


Figure 11 (left): Transmission of tungsten (W) collimators as a function of gamma energy. Figure 12 (right): MCNP simulation of the background caused by Compton scattering in a 4" tungsten collimator with a conical aperture. Only high-energy gammas undergo multiple large-angle scattering events to produce a low-energy background.

3. Instrument Operation

The Ge detector is cooled to its operating temperature in a liquid nitrogen dewar and biased at +600 V (figure 13). The source is held at a distance of ~8" in front of the W shield at a distance of ~14" to the Ge detector. The source is surrounded by a shield made out of 2" Pb bricks (not shown) to reduce scattering from the rest of the instrument and the laboratory. The shield is open in the back to avoid direct backscatter into the Ge detector (cf. figure 2). Gamma signals are amplified with a custom-built charge sensitive preamplifier with an FET input noise of ~1 nV/√Hz at room temperature and a feedback capacitance of 0.5 pF. The primary veto consists of a single 3.5" x 6" CsI crystal, while the secondary annular veto consists of 8 separate CsI wedges with individual PMTs whose signals are summed into a single output. The PMTs of the two vetoes are biased at +1000 V, and the three output signals (Ge, CsI1 and CsI2) are recorded in a 4-channel 100 MHz 16-bit Pixie-4 digitizer from XIA. The signals are shaped with a trapezoidal digital filter whose time constants of order ~1 µs are set shorter than required for highest energy resolution to reduce pile-up and increase the fraction of vetoed events. Trigger levels are set just above the noise to enable vetoing even for small energy deposition in the scintillators. The anti-coincidence logic is implemented in software.

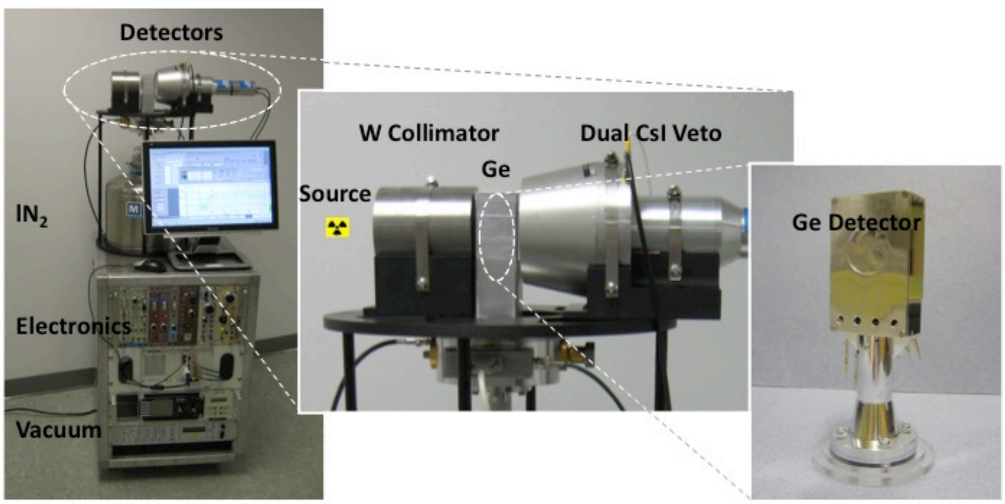
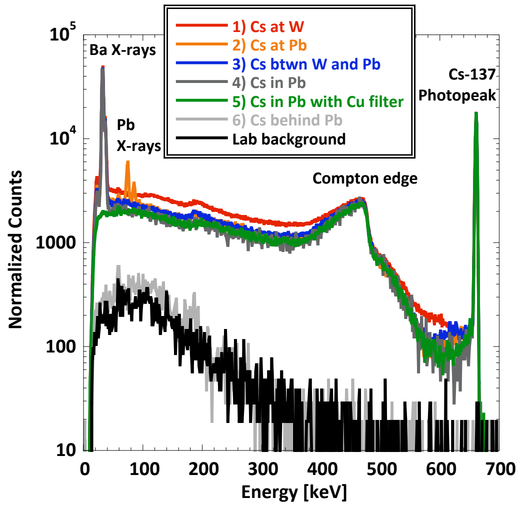


Figure 13: Photographs of the Ge detector with the W shield in front of it and the two CsI Compton vetoes behind it. The Pb source shield in front of the W collimator is not shown in this configuration.

4. Results

4.1. Geometry Optimization

It is important to appreciate that the low-energy background can only be suppressed by if it is caused by forward scattering *in* the Ge detector. The fraction of photons that scatter to low energy *before* they reach the Ge and then deposit their energy without leaving a signal in the vetoes cannot be suppressed. The geometry of the shield and the collimator must therefore be optimized, so that photons are either fully absorbed, or bypass any shielding (and other masses nearby) completely. The difference in the Compton background caused by a pure Cs-137 source for different geometries is illustrated in figures 14 and 15. If the source is held close to the end of the W collimator, a large fraction of the gammas will illuminate its inside and increase the Compton background. Photons that are scattered *once* in a small angle from the inside of the W collimator into the Ge detector produce the increase between 550 and 650 keV, while multiple scattering in the collimator produces the increase at lower energy (red trace). This contribution can be reduced by moving the source *away* from the collimator along the line of sight.



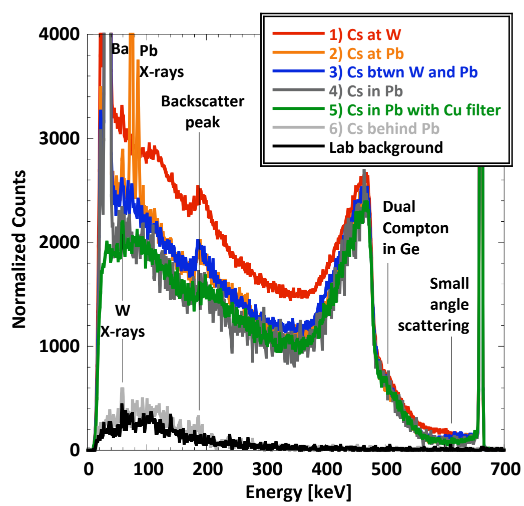


Figure 14 (left): Compton background for different source and shielding geometries. All Cs-137 spectra are scaled to 10^5 total counts in the photopeak. Figure 15 (right) shows the same data on a linear scale.

To first order, the extent to which this contribution to the Compton background cab be reduced depends on the ratio of the solid angle for illuminating the inside of the W collimator and for illuminating the Ge detector directly. This means that placing the source close to the Pb shield or between the Pb and the W collimator still adds background to the spectra that cannot be suppressed (orange and blue spectra). In addition, if the source is held too closely to the Pb shield, the characteristic lines of the shielding materials appear in the spectra. The strong Pb lines in this case (orange spectrum) reflect the fact that X-ray fluorescence is emitted isotropically, while Compton scattering is forward-peaked.

The best location for the source is *inside* a Pb shield that is large enough so that the source can be kept at least a few inches away from any of the walls. This ensures none of the radiation emanating from the shield has a direct line of sight to the Ge detector. Our current shielding uses 2" thick Pb bricks and encloses a total volume of 4" x 4" x 8", with the source suspended by thin wires in the center. This is sufficient to reduce the Compton background close to its limiting value (dark grey spectrum), although both shield thickness and volume should be increased in future designs. As outlined in figure 2, the Pb shield should be open in the back with a significant distance between the source and the laboratory wall so that gammas emitted along the line of sight in opposite direction to the Ge detector have a small probability of backscattering into it. For some samples it may be desirable to suppress the low-energy X-rays from the source, not because they fall into the ~100 keV region of interest for Pu analysis, but because they have a high intensity and will therefore increase the dead time and reduce the veto efficiency of the detector. This can be done with a ~1 mm Cu filter at the *midpoint* between source and detector (green spectrum), so that any small-angle scattering produces gammas above the ~100 keV region of interest, and any large-angle scattering will not reach the Ge detector.

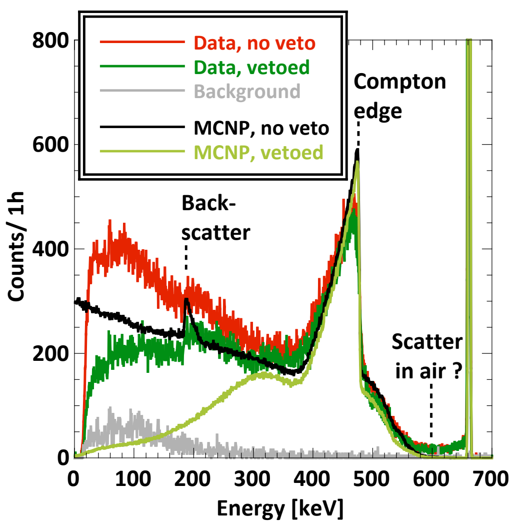
Since the Ge detector and the Compton vetoes are not enclosed in any shielding in the current setup, radiation from (mostly) the natural radioactivity in the concrete walls of the laboratory will contribute to the spectral background (black spectrum, scaled to the same data acquisition time of 1h as the green and grey spectra with the source inside the Pb shield). This produces a limiting background for these experiments, although it could be suppressed in the future. Note that enclosing the source *inside* the Pb shield is important, because scatter from the source in the laboratory will contribute to the background even if the direct line of sight is blocked by 8" of Pb (light grey spectrum).

4.2. Compton Veto

When comparing the measured spectra for the optimized geometry with Monte Carlo simulations, it is evident that the Compton background agrees qualitatively, but not quantitatively even in the case without a Compton veto (figure 16). The laboratory background alone with all the sources removed cannot fully account for this discrepancy. Rather, it is partially due to simplifications in our current Monte-Carlo models, and partially due to non-idealities in the experimental setup.

Our MCNP simulations currently only include the main parts of the instrument (shield, collimator, cryostat, windows etc.), while other parts that contribute to the background (dead layers in the Ge detector, PMTs, scintillator cases, electronics rack, other equipment, laboratory walls, air etc.) are neglected for simplicity. The finite size of the Cs-137 source is also not included here to see how closely our setup approaches the ideal case of negligible in-source scattering. In addition, MCNP does not keep track of electrons below 1 MeV to limit computation time, and thus does not account for electron escape that increases the background at the expense of the full photopeak at 662 keV. The finite background between ~550 keV and ~650 keV, which is directly proportional to the number of electrons between the source and the detector, suggests that scattering in air may also contribute. Finally, MCNP assumes Compton scattering off free electrons, while electrons are bound to the Ge atoms in the detector with a finite binding energy of up to ~10 keV. This broadens the Compton edge at ~470 keV. Since the discrepancy between data and MCNP below ~180 keV roughly follows the shape of the laboratory background, we believe that the dominant discrepancy between data and simulations is caused by multiple scattering of the source gammas in the instrument and the laboratory environment.

The primary limitations of the current setup are the straight (rather than conical) shape of the W collimator and the finite volume of the 2" Pb shield. Both effects lead to an unnecessary increase in the areas where a single or two successive scattering events can produce a low-energy photon with a direct line-of-sight with the Ge detector (cf. figure 2). The finite Pb thickness also increases the illumination of the rest of the instrument. These effects can be reduced with minor modifications in the spectrometer design.



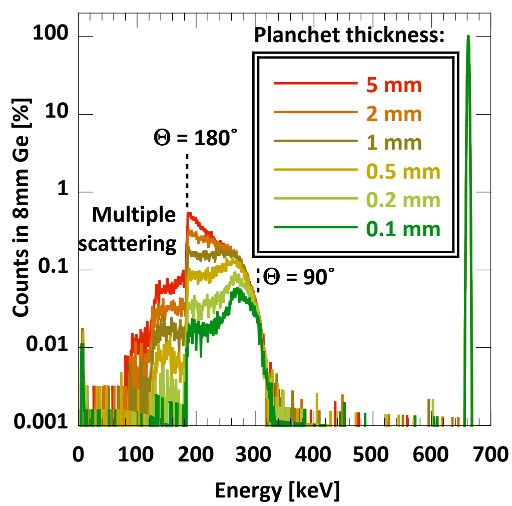


Figure 16 (left): Low-energy background suppression in the Ge detector by anti-coincidence vetoing with the two CsI scintillator vetoes. Simple MCNP simulations (black and light green) and a background spectrum (grey) are included for comparison. Figure 17 (right): In-source scattering from the 0.5 mm thick 1/2" diameter stainless steel planchet that supports the radioactive Cs-137 source increases the background in the energy range between ~180 and ~300 keV.

We can repeat the experiment with the two CsI scintillators activated and reject all events from the Ge detector that leave a coincident signal in one of the vetoes. As expected, the background below 300 keV is now reduced, because it is mostly caused by gamma-rays that are forward-scattered in the Ge detector. We observe the strongest background reduction in the energy range around ~100 keV where the Pu X-ray and gamma lines of interest are. This successfully demonstrates the approach to selectively suppress the low-energy background with a Compton veto placed *behind* the primary Ge detector.

However, the degree of reduction is not quite as high as predicted by MCNP. This is primarily due to excess low-energy background that was already evident in the spectrum without a veto. Since these events are caused by scattering outside the Ge detector and do not produce a signal in the CsI scintillators, they can obviously not be rejected by anti-coincidence vetoing. Interestingly, the backscatter peak does not fully disappear as predicted by MCNP. This is because MCNP only includes backscatter from the CsI scintillators (which always leaves a signal in them), but not from the source itself. Despite the small source size of 0.5 mm thickness and 1/2" diameter, in-source scattering produces a noticeable background in the energy range between ~150 and ~300 keV (figure 17). This background has a different shape than the typical backscatter peak, because it is not caused mostly by ~180˚ scattering, but by scattering angles over the full range between 180˚ and 90˚.

5. Summary and Outlook

This work successfully demonstrates the suppression of the low-energy gamma background with a Compton veto placed directly *behind* a *thin* Ge detector. In the ideal case, the degree of suppression depends on the solid angle coverage and the quantum efficiency of the Compton veto used, and can be several orders of magnitude. In practice, scattering anywhere other than in the active detector volumes can contribute a certain fraction to the background that cannot be removed by anti-coincidence vetoing. This includes scattering in the source itself, the source shielding, collimators, the cryostat and any other part of the experiment and the surrounding laboratory. It is therefore essential to optimize all aspects of the geometry, the most important of which are:

1) Make the source as small as possible (a jet of micro-droplets would be ideal)
2) Collimating the source onto the detector center (conical collimator, half way between them)
3) Minimize all mass in the line of sight between source and detector (including behind the source)
4) Maximize shielding in all other directions around the source (and, ideally, the detector)

In this work we have implemented the fist steps in this optimization process. We have shown that the instrument works qualitatively as expected, although there are still quantitative differences between the experimental data and Monte-Carlo simulations. These differences are likely due to multiple-scattering events in the material surrounding source and detector, and a non-optimal collimation.

What is needed now, in addition to some obvious improvements in the collimator and shield geometry, is a *quantitative* accounting of *all* different scattering processes in the surrounding material that contribute to the Compton background. Only then will it be possible to predict by how much the Compton background can be suppressed with an anti-coincidence veto, and how much of it is due to unavoidable scattering in essential surrounding matter. Once this limit is known quantitatively, it will be possible to predict how much further the Compton background can be suppressed with bigger and more efficient BGO vetoes (which are commercially available), and if this suppression can be sufficient to directly detect Pu emissions from spent nuclear fuel. This will be the subject of a future proposal.